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Conduction Band States and 5d-to-4f Laser Transition of Rare Earth Ion Dopants

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Since the 5d-4f transition of CaF_2:Sm^{2+} was demonstrated to lase many years ago, researchers have sought to take advantage of the high cross sections and broad bandwidths afforded by these types of transitions. It was realized early-on that the 5d-4f transition suffered from numerous potential drawbacks, however, including nonradiative decay, excited state absorption, and solarization of the host medium. A closer look at the problem reveals that all these issues can be linked to the interaction of the conduction band states with the 5d orbitals of the rare earth ion.

A few years ago we sought to understand why the 4f-5d transition of the Sm^{2+} ion lases in the CaF_2 host, while Eu^{2+} has no gain. An extensive investigation of the pump-induced gain/loss throughout the UV-visible-IR showed that *both* Sm^{2+} and Eu^{2+} exhibit broad, intense excited state absorption (ESA) features that extend from about 700 to 300 nm in CaF_2, missing the 750 nm emission region for Sm but overlapping it for Eu (near 430 nm). The nature of the ESA was confirmed by recording these spectra for Eu^{2+} in the fluorite-structure series CaF_2-SrF_2-BaF_2-$SrCl_2$, and noticing that the ESA peak is strongly correlated with the relative position of the conduction band (CB) and the impurity energy levels, as theoretically described by McClure and others.

The nonradiative decay experienced by Sm^{2+} can also be attributed to the interaction of the 5d orbital with CB states. This issue was examined by comparing the Sm nonradiative decay rates in numerous fluorite-structure crystals and in fluoride glasses. In this way it was possible to develop a picture where thermal activation of an electron from the 5d level to the CB could be identified as the first step of the nonradiative pathway. In fact the emission quenching could be related to the CB position as deduced from the ESA data for the CaF_2-SrF_2-BaF_2-$SrCl_2$ series. The specific composition of different fluoride glasses also had significant impact on the Sm^{2+} emission efficiency, in that the glasses consisting of more ionic components had much higher emission yields.

Ce^{3+} ions have long been known to exhibit laser action in the UV region, although the laser performance is impaired by both ESA and solarization of the host. The early efforts have concentrated on such hosts as $LiYF_4$ and LaF_3, but Dubinskii and others recently showed that $LiCaAlF_6$ and $LiSrAlF_6$ provide the most robust laser operation observed to date with a laser based on the 5d-4f transition. The key to the favorable performance appears to be the fundamental anisotropy of the CB in these crystals, in that the 5d-CB transition is strongly polarized with E perpendicular to the c axis, (so the laser is efficient when operated in the orthogonal polarization). A certain amount of solarization also occurs, although it exhibits the same polarization dependence as the ESA and can be minimized by including the appropriate codopants in the crystal along with the cerium.

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